

Particle Emissions and the Production of
Ozone and Nitrogen Oxides from the
Burning of Forest Slash

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PARTICLE EMISSIONS AND THE PRODUCTION OF OZONE AND NITROGEN OXIDES FROM THE BURNING OF FOREST SLASH

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Abstract - Airborne measurements in the plumes from three prescribed burns of conifer slash showed the number concentration-size spectra to be bimodal with peaks at ~ 0.1 and $\sim 0.5 \mu\text{m}$. The mass distribution was sharply peaked at $\sim 0.3 \mu\text{m}$ where over 80% of the mass of the particles in the plume resided. Most of the particles in the plumes were of primary rather than secondary origin. The burns emitted particulate mass into the atmosphere at rates of $0.1\text{--}15 \text{ kg s}^{-1}$ during the active burning phase. The average density of the particles ranged from 0.75 to 1.34 g cm^{-3} . Estimates of particle emission factors for the burns ranged from 0.2 to 2%, higher values being associated with higher fuel consumption rates.

The burns were prolific sources of cloud condensation nuclei (CCN), producing $\sim 10^{10}\text{--}10^{11}$ CCN per gram of wood consumed. The CCN resulted in anomalously high concentrations of water droplets $< 10 \mu\text{m}$ diameter in the cumulus clouds produced by the burns.

Ozone concentrations near the tops of the plumes reached values as high as 44 ppb above ambient values, the higher values generally being associated with high u.v. intensities. Peak concentrations of NO_2 and NO in the plumes were ~ 60 ppb; the ratio of NO_2/NO ranged from 1 to 3. SO_2 was not found in the plumes.

1. BACKGROUND

In a previous paper (Radke *et al.*, 1978) we reported on airborne measurements made in the emissions from several prescribed burns of forest slash in Washington State. Among the major features of these observations were a surprising consistency in the particle size distributions from one burn to another, a suggestion that previous estimates of the amount of suspended particulate per unit mass of wood consumed may be too low, and indications that some of the burns were substantial sources of ozone. However, these observations remained tentative due to the very limited data set.

In this paper we present the results of a more detailed study of the particles and trace gases produced by three prescribed burns in Washington and Oregon. The measurements were obtained with the University of Washington's B-23 research aircraft (Hobbs *et al.*, 1979) in concert with surface observations by foresters.

2. DESCRIPTION OF THE BURN

The first burn studied occurred on 19 July 1978 about 130 km southwest of Seattle, Washington and covered an area of 0.63 km^2 . The burn was started with diesel oil and gasoline from a drip bucket aboard a helicopter. Lighting began at 1405 Pacific Daylight Time (PDT) and continued until 1520 PDT. The burn

was well developed by 1450 and started to die out by 1620 PDT. During the vigorous burning period a cloud formed near the top of the plume about 2 km downwind; most of the plume passed beneath this cloud. The estimated fuel consumed by the burn was $\sim 5.6 \times 10^6 \text{ kg}$ conifer slash, with an estimated fuel moisture content of $\sim 10\%$. (The fuel parameters reported in this paper were not measured precisely but obtained from estimates made by foresters.)

The second burn occurred on 22 September 1978 about 50 km south of Eugene, Oregon, and covered an area of 0.34 km^2 . This burn was hand-lit, starting about 1110 PDT; lighting continued somewhat intermittently until 1500 PDT. By 1630 PDT the burn entered a smoldering stage. Due perhaps to the comparatively uneven lighting sequence, this burn was much less steady-state than that on 19 July and it produced several periods of intense burning that created large cumulus clouds. The plume was almost entirely engulfed by these clouds during the periods of intense burning. As a result, the samples that were obtained in cloud-free air were from the less intense burning periods. The burn consumed an estimated $3 \times 10^6 \text{ kg}$ of conifer slash, with $\sim 12\%$ moisture content.

The third burn occurred on 5 October 1978 about 50 km southeast of Olympia, Washington and covered an area of 1.13 km^2 . Hand-lighting of the burn began about 1100 PDT and continued until 1200 PDT. A helicopter with a drip bucket began lighting the fuel at 1245 PDT and continued until 1545 PDT. The fuel moisture ($\sim 20\%$) was higher than the other two burns; the burn did not develop as rapidly but it was

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by far the most steady-state of the three burns studied. During the period 1200–1530 PDT the burn produced a steady and rather small plume. During the period 1530–1700 PDT the burn was more involved, resulting in a much larger but still fairly steady plume. During the latter period there was a small secondary plume, produced by some areas of smoldering combustion, which was separate from the main plume. The burn eventually consumed $\sim 5 \times 10^6$ kg conifer slash.

3. PARTICLES IN THE PLUMES

Particle number distributions measured in each of the burns are shown in Fig. 1. The particle size distributions measured in the plumes from each of the burns were similar in shape, with a major peak at $\sim 0.1 \mu\text{m}$ diameter and a smaller peak at $\sim 0.5 \mu\text{m}$; the concentrations of particles of a given size were also remarkably similar from one burn to another. An example of the particle volume distributions is shown in Fig. 2. The peak in the volume distribution at $\sim 0.3 \mu\text{m}$ dia. corresponds to the peak at $\sim 0.1 \mu\text{m}$ in the number distribution. Generally, over 80% of the volume (or mass) of the particles was contained in this peak. A similar peak in the volume distribution (determined from an optical inversion technique) was observed by Veretennikov *et al.* (1979) for wood smoke.

It can be seen from Figs 1c and d that there was little change in the particle size distribution with range, with the possible exception of slightly lower small particle concentrations at greater distances downwind (which may reflect the effects of particle coagulation). Since the changes in the sizes of the particles with travel time are small, the majority of the particles in the plumes must have originated as primary particles from the burn itself, rather than as secondary products produced in the plumes. In this respect, the plumes produced by the burning of forest slash differ significantly from those from many industrial processes, such as the plumes from coal-fired power plants (Forrest and Newman, 1977; Gillani *et al.*, 1978). Packham and Vines (1978) have suggested, on the basis of their measurements of Aitken nuclei and light scattering coefficients in several prescribed fires, that there is little coagulation of smoke particles once they have formed. Our measurements of the particle size distributions support this suggestion.

The densities of the particles in the plumes from the burns were computed by comparing the measured mass concentrations of the particles with the total volume concentrations of particles in the plumes. The results are shown in Fig. 3, where the derived particle mass concentrations are plotted against the particle volume concentrations. The slope of the regression line, which was fitted to each of the data sets, gives the average density of the particles in the plume. The average particle densities for the burns on 19 July, 22 September, and 5 October were 0.75, 0.94 and 1.34 g cm^{-3} , respectively. It is interesting that these densities

correlate with the fuel moisture contents.

Cloud condensation nuclei (CCN) (active at 0.2% supersaturation) measured in the plumes ranged from 2000 to 8000 cm^{-3} . Concentrations in the ambient air were generally less than 100 cm^{-3} . The concentrations of particles larger than $0.1 \mu\text{m}$ (which can act as CCN even if they contain relatively small amounts of water-soluble materials) were high enough to explain the observed CCN concentrations. The rates of emission of CCN from the burns were on the order of 10^{16} – 10^{17} s^{-1} , which places them on a par with very large urban areas and the most prolific industrial sources of CCN (Radke and Hobbs, 1976). From our airborne measurements of CCN concentrations in the plumes and the estimated rates of burning of fuel, we deduce that $\sim 10^{10}$ – 10^{11} CCN were produced per gram of wood burned.

The burns on 19 July and on 22 September produced cumulus clouds of considerable size. Both clouds ingested portions of the plume. The measured cloud droplet size distributions in these clouds are shown in Fig. 4. The clouds contained extremely high concentrations of small ($< 10 \mu\text{m}$) droplets compared to the ambient clouds (the concentrations of small droplets in similar ambient clouds were more than an order of magnitude less than those shown in Fig. 4). We attribute this to the very high concentrations of CCN in the plumes.

4. PARTICLE EMISSION RATES AND EMISSION FACTORS

From the measurements of the concentrations of particles in vertical cross-sections downwind of the burns, differences between the average concentrations of particles in the plumes and in the ambient air were deduced. Combination of these results with the horizontal winds and cross-sectional areas of the plumes yields the total flow of particles in the plumes. This approach is limited to steady-state plumes and is probably accurate to only about $\pm 20\%$.

From the measurements made 10 km downwind in the plume on 19 July, we calculated that the burn was producing particulate mass at a rate of $\sim 15 \text{ kg s}^{-1}$ and CCN (active at 0.2% supersaturation) at a rate of $\sim 1.4 \times 10^{17} \text{ s}^{-1}$. Since the emissions that were sampled at that range were emitted during the most vigorous burning period, these estimates are probably representative of the maximum rates.

Since the burn on 5 October was in a much more steady-state condition than the other two burns studied, it is possible in this case to estimate the particle production rate for various distances downwind and therefore for various stages of the burn. During the milder stages (1230–1530 PDT) of the burn on 5 October, the rates of production of particulate mass were estimated from measurements made at 1.8, 9.3 and 18.5 km downwind to be 0.5, 0.1 and 0.4 kg s^{-1} . During the most vigorous burning stage

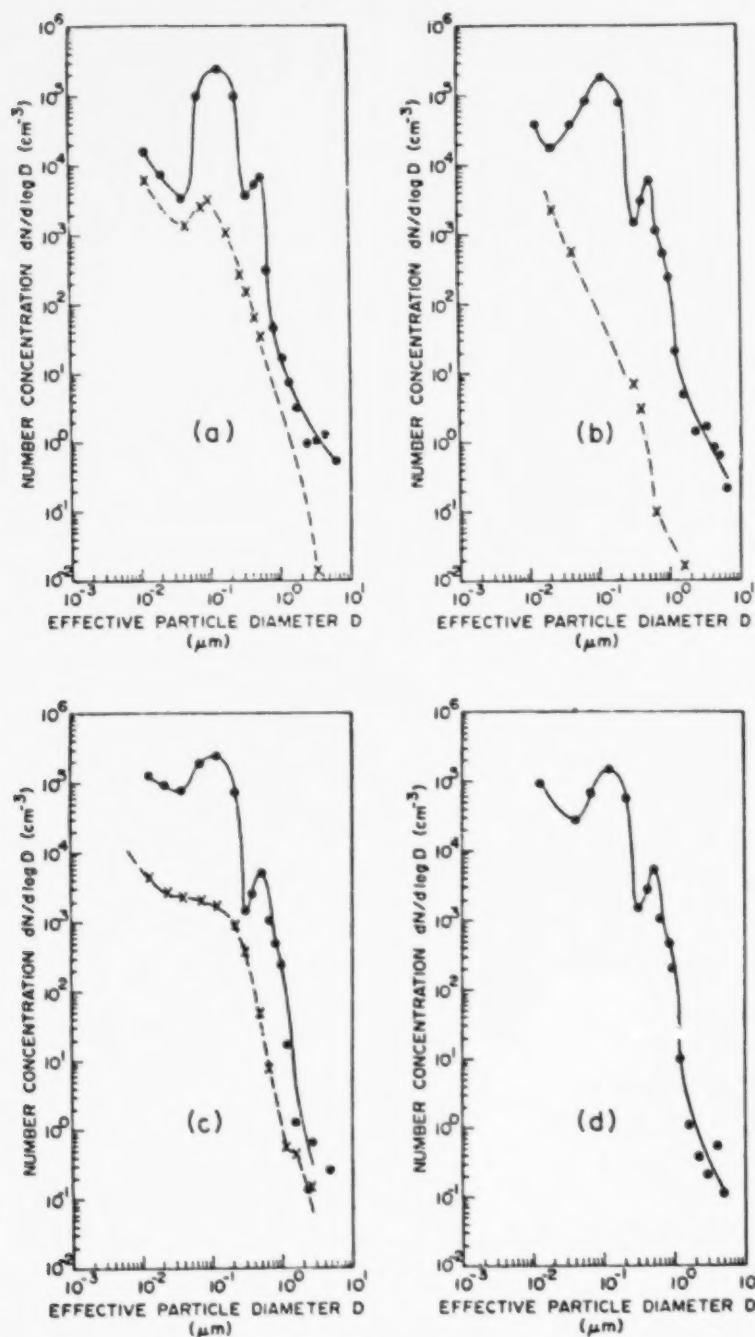


Fig. 1. Number concentration vs size of particles measured in the plumes from the prescribed burning of forest slash on: (a) 19 July 1978 at an altitude of 1829 m (MSL) and 10 km downwind of the burn; (b) 22 September 1978 at an altitude of 1981 m and 7.4 km downwind; (c) 5 October 1978 at an altitude of 1402 m and 1.8 km downwind; and (d) 5 October 1978 at an altitude of 1219 m and 18.5 km downwind. The dashed curves show the ambient values.

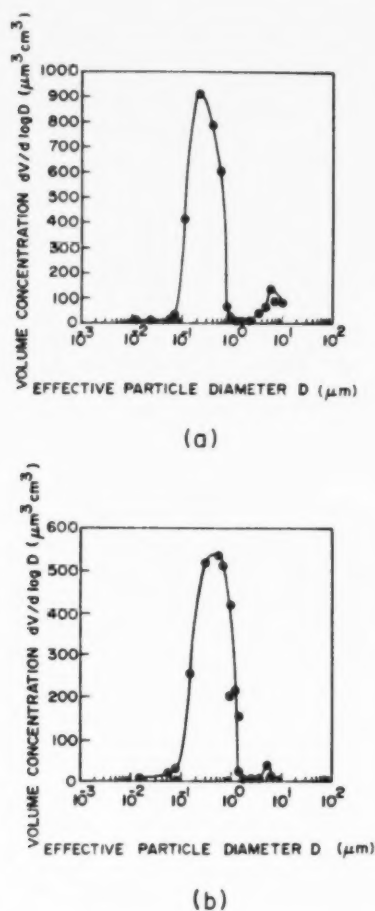


Fig. 2. Volume concentration vs size of particles measured in the plumes from the prescribed burning of forest slash on: (a) 19 July 1978 at an altitude of 1829 m (MSL) and 10 km downwind of the burn; and (b) 5 October 1978 at an altitude of 1219 m (MSL) and 18.5 km downwind of the burn.

(1530–1700 PDT) the production rates of particulate mass estimated from the cross-sectional measurements made at 1.8 and 9.3 km downwind were 1.8 and 1.3 kg s^{-1} , respectively. The CCN emission rate during the milder burning stage was $\sim 9 \times 10^{15} \text{ s}^{-1}$ (active at 0.2% supersaturation), and during the more vigorous period it was $\sim 6 \times 10^{16} \text{ s}^{-1}$.

According to ground-based observations made on 19 July, about 75% of the fuel was consumed between 1430–1600 PDT. This gives an average fuel consumption rate for this period of $\sim 8 \times 10^2 \text{ kg s}^{-1}$. Since the fire was producing particulate mass at a rate of $\sim 15 \text{ kg s}^{-1}$, the emission factor (i.e. mass of particles produced per unit mass of fuel consumed) was $\sim 2\%$. For the burn on 5 October, ground-based observations indicate that $\sim 40\%$ of the fuel was consumed during the milder burning period, giving an average fuel consumption rate of $\sim 1.9 \times 10^2 \text{ kg s}^{-1}$. Since the burn on 5 October was producing

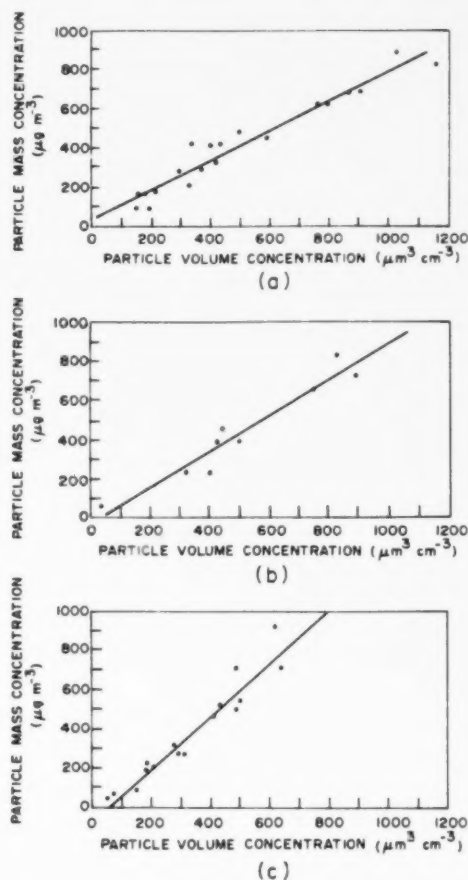


Fig. 3. Mass concentration of particles vs total particle volume concentration for particles collected in the plumes from the prescribed burning of forest slash on: (a) 19 July 1978; (b) 22 September 1978; and (c) 5 October 1978.

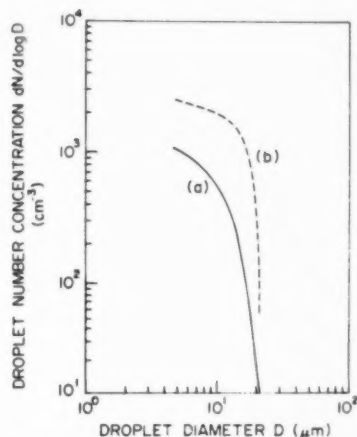


Fig. 4. Cloud droplet size distributions measured in clouds created by the prescribed burning of forest slash on: (a) 19 July 1978 at an altitude of 2987 m (MSL) and 1.8 km downwind of the burn; and (b) 22 September 1978 at an altitude of 2591 m and 9.3 km downwind of the burn.

particulate mass at an average rate of $\sim 0.3 \text{ kg s}^{-1}$ during this period, the emission factor was $\sim 0.2\%$ during the milder burning stage. During the vigorous burning period $\sim 40\%$ of the fuel was consumed. The remaining 20% of the fuel was consumed during the startup and smoldering stages of the burn. During the vigorous burning period, the average fuel consumption rate was $\sim 3.7 \times 10^2 \text{ kg s}^{-1}$, giving an emission factor of $\sim 0.4\%$.

One of the most subjective elements in estimating emission factors in this way is in the determination of the total amount and rate of burning of the fuel; as mentioned previously, in our studies these were estimated by foresters. A promising alternative method is to measure the concentration of CO_2 in the plume. A preliminary attempt was made to obtain such measurements from our research aircraft by filling previously evacuated cylinders with samples from the plume and then analyzing the sample for CO_2 content by gas chromatography. The measurements at 9.3 km downwind on 5 October yielded a CO_2 emission rate of $\sim 430 \text{ kg s}^{-1}$. If we assume that the fuel had a carbon content of $\sim 44\%$ by weight, the rate of fuel consumption would have been $\sim 267 \text{ kg s}^{-1}$. This compares favorably with our previous estimate of $\sim 190 \text{ kg s}^{-1}$ for the same period.

Our results suggest that there is considerable variability in emission factors for one burn to another and that the emission factor may be a function of the fuel consumption rate. Although we have measurements on only three burns, the more rapidly the fuel was consumed the higher the emission factor. If this result proved to be generally true, it would probably be appropriate to use a higher emission factor for wildfires than for most prescribed burns of forest or agricultural products. The conditions present on the 19 July burn probably come closest to wildfire conditions, since the fuel was consumed quite rapidly. However, unlike most wildfires, none of the burns we studied reached the intense "crown fire" stage where the fire is burning in the upper portion of standing trees.

5. PLUME STRUCTURE AND VISIBILITY

Cross sections of the light scattering coefficients b_s are presented in Fig. 5 for the 19 July burn and in Figs 6 and 7 for the milder burning period and the more vigorous burning period, respectively, on 5 October. The plume on 5 October consisted of two main lobes near to the fire, one above the other (Figs 6a, 7a). Further downwind these lobes merged into one (Figs 6b, c, 7b). This plume behavior was observed both during the morning (the milder burning period) and during the afternoon (the vigorous burning period). A very similar plume behavior is observed in the study by Packham and Vines (1978, Fig. 1). Two lobes are also evident in the structure of the 19 July fire (Fig. 5). Evidently, the existence of two lobes, one above the other, is a recurring feature of plumes close into fires;

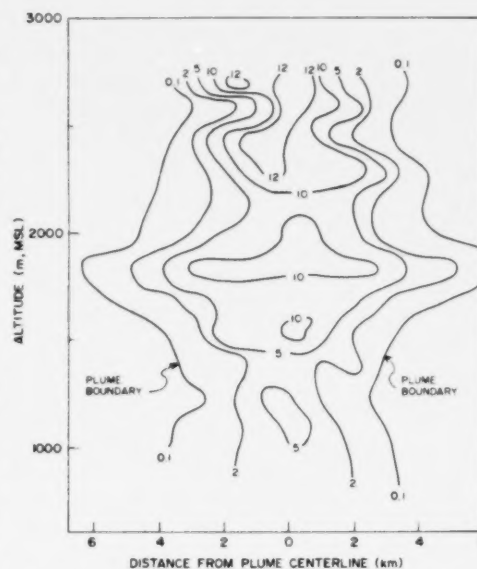
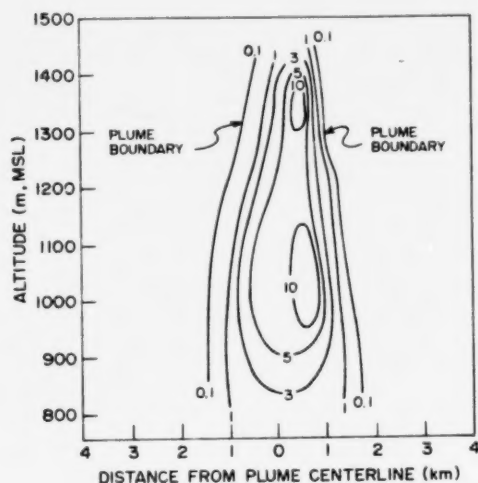


Fig. 5. A vertical cross-section of the light-scattering coefficient (in units of 10^{-4} m^{-1}) measured perpendicular to the long axis of the plume at 10 km downwind from the prescribed burn at 1522-1615 PDT on 19 July 1978. The height of the terrain was between sea level and 200 m (MSL).

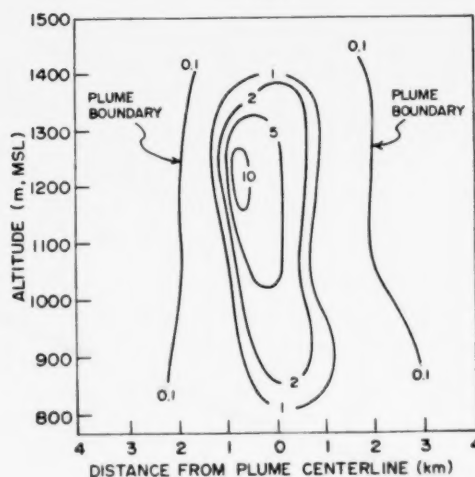
further downwind these lobes merge into one. The lobes may be due to different temperature regions within the fire (e.g. active burning and smoldering areas).

The peak value of b_s for the burn on 19 July was $\sim 12 \times 10^{-4} \text{ m}^{-1}$ (Fig. 5) which, neglecting the effects of absorption of light by particles and gases, corresponds to a visual range of $\sim 3.3 \text{ km}$ (the visual range in the ambient air was $\sim 130 \text{ km}$). The peak value of b_s for the plume from the milder burning period on 5 October was $\sim 10 \times 10^{-4} \text{ m}^{-1}$ (Figs 6a, b) which corresponds to a visual range of 3.9 km. During the more vigorous burning period on 5 October, the peak value of b_s was $\sim 30 \times 10^{-4} \text{ m}^{-1}$, which corresponds to a visual range of only 1.3 km. However, this value was only found in certain localized regions of the plume which resulted from the non-steady state nature of the source (Fig. 7c).

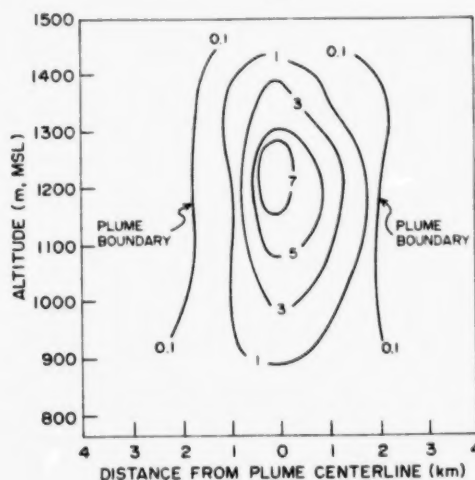
The ratio of the mass concentration of particles to the light-scattering coefficient in the plumes was $0.75 \pm 0.28 \text{ g m}^{-2}$ on 19 July and $0.17 \pm 0.05 \text{ g m}^{-2}$ on 5 October. The higher value of this ratio on 19 July was probably due to the fact that a larger fraction of the total mass was contributed by particles $> 1 \mu\text{m}$ in dia. (which do not contribute significantly to b_s) in the plume on 19 July than in the plume on 5 October. In general, the particles from the burning of forest products are very effective as light-scatterers due to the fact that most of their mass is contained in particles a few tenths of a micrometer in dia. (Fig. 2), which lie within the size range for the most effective scattering of light.



(a)



(b)



(c)

6. OZONE AND NITROGEN OXIDE CONCENTRATIONS

The measured concentrations of ozone in the plume from the burns were often greater than in the ambient air. However, on some occasions the ozone concentrations in the plumes were less or equal to those in the ambient air. An example of some of the ozone data is shown in Fig. 8. A more complete listing of the ozone, particle mass loading, and the u.v. flux data is presented in Table 1.

Measurements of ozone concentrations 10 km downwind of the burn on 19 July are shown in Fig. 9. Maximum ozone concentrations of ~140–120 ppb (~68–58 ppb above ambient values) were present near the middle and top of the plume with an ozone deficit of ~25 ppb near the base of the plume. The 22 September 1978 fire, which produced an intermingled plume of smoke and cumulus clouds, produced little or no ozone during our limited observations. It is not clear whether the cloud simply acted to reduce the ultraviolet intensity (as in the samples of 1314, 1318 and 1334 PDT) or the presence of droplets inhibited the formation of ozone (as at 1324 and 1405 PDT). The ozone measurements obtained on 5 October are depicted in Fig. 10. The peak ozone concentrations in the plume were 100 ppb (~27 ppb above ambient) at downwind distances of 15–20 km; the peak values again occurred near the middle and the top of the plume. Two of the passes (at 1550 and 1629 PDT) close into the burn on 5 October showed considerable ozone deficits. In these regions the u.v. intensity was reduced by extremely high particle concentrations (see Table 1) and the ozone may have reacted with the particles. The general level of the ozone concentrations was somewhat lower on 5 October than on 19 July, as was the u.v. intensity.

It can be seen from Figs 11a and b that, after a few kilometers of travel, the excess ozone concentrations in the plumes correlated well with the u.v. fluxes on 19 July and 22 September (correlation coefficients 0.90 and 0.93, respectively). However, on 5 October there was no such correlation (Fig. 11c). In this case, the measurements divide into two groups: those taken prior to 1530 PDT when the plume was rather small and the ozone levels comparatively low even though the u.v. flux was high, and those taken after 1530 PDT when the plume was much larger and the ozone levels generally higher even though the u.v. flux was low.

Fig. 6. Vertical cross-sections of the light-scattering coefficient b_s (in units of 10^{-4} m^{-1}) measured perpendicular to the long axis of the plume from the prescribed burn on 5 October 1978 during the period 1202–1309 PDT: (a) 1.8 km downwind, (b) 9.3 km downwind and (c) 18.5 km downwind. The height of the terrain was 200–500 m (MSL).

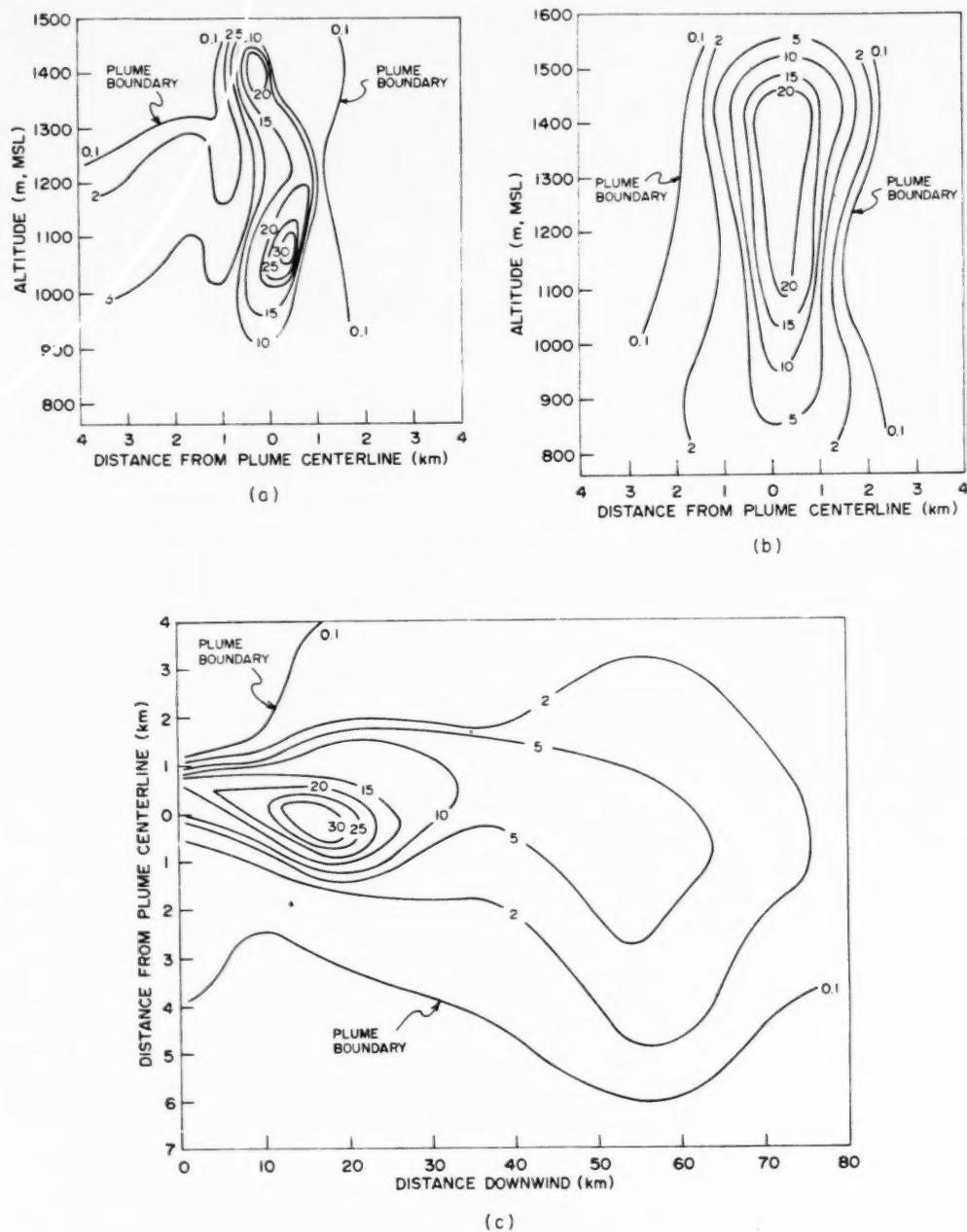


Fig. 7. Vertical cross-sections of the light-scattering coefficient b_s (in units of 10^{-4} m^{-1}) measured perpendicular to the long axis of the plume from the prescribed burn on 5 October 1978 during the period 1543–1613 PDT: (a) 1.8 km downwind, and (b) 9.3 km downwind. A plan view of the light scattering coefficient (in units of 10^{-4} m^{-1}) at altitudes of 1158–1780 m in the plume for the period 1539–1709 PDT is shown in (c). The height of the terrain was between 200–500 m (MSL).

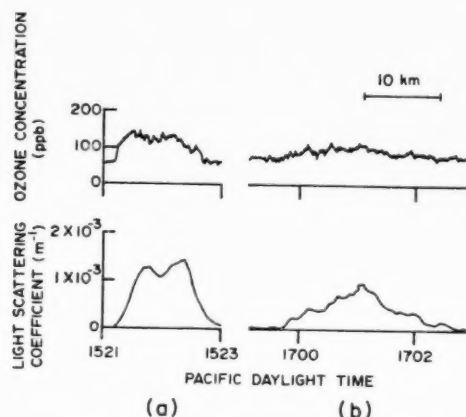


Fig. 8. Ozone concentrations and light-scattering coefficients measured in the plumes from the prescribed burning of forest slash on: (a) 19 July 1978, near the top of the plume at an altitude of 2682 m (MSL) and 10 km downwind; and (b) 5 October 1978, near the center of the plume at an altitude of 1280 m and 55.6 km downwind.

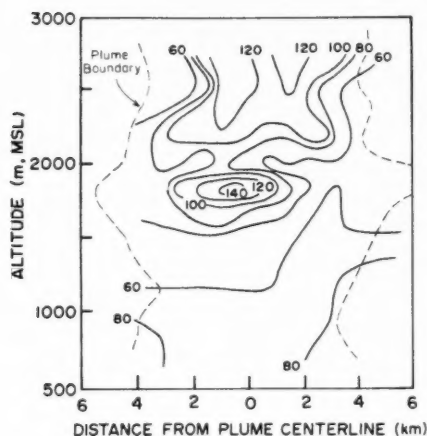


Fig. 9. A vertical cross-section of the ozone concentrations (in ppb) measured perpendicular to the long axis of the plume from the prescribed burn on 19 July 1978 at 10 km downwind. The concentration of ozone in ambient air ranged from 85 ppb at 1200 m to about 62 ppb at 2600 m altitude. The height of the terrain varied from near sea level to 200 m (MSL).

The minimum concentrations of NO_2 and NO that could be detected with the instruments aboard the aircraft were ~ 20 ppb. The concentrations of these two gases in the ambient air unaffected by the plumes from the burns never reached this value. Peak concentrations of NO_2 and NO of ~ 60 ppb were measured in the plume from the 19 July burn at 3 km downwind, and concentrations of NO_2 and NO in the plumes were only just above 20 ppb at ranges beyond 10 km. The measured concentrations of NO_2 and NO_x in the plume from the 5 October burn were only just

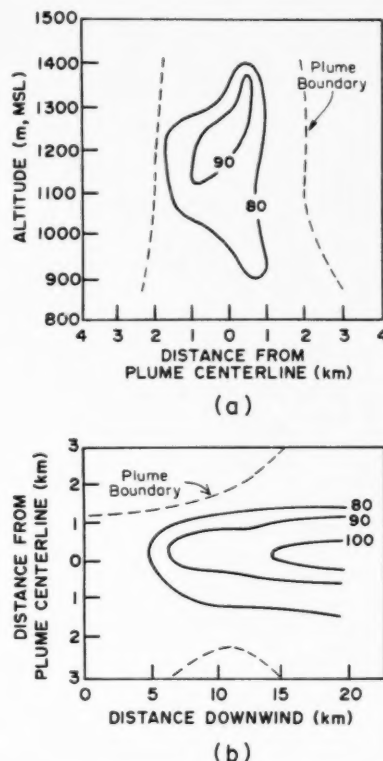


Fig. 10. Ozone concentrations (in ppb) measured in the plume from the prescribed burn on 5 October between 1153 and 1309 PDT: (a) in a vertical cross-section 9.3 km downwind of the burn, and (b) in plan view at an altitude of 1219 m. The concentration of ozone in the ambient air was about 73 ppb. In (a) the terrain was hilly, averaging 500 m (MSL). In (b) the terrain was hilly with average elevations decreasing from 600 to 100 m (MSL) with increasing range.

above 20 ppb at all ranges.

The ratio of NO_x/NO in the plumes ranged between 1 and 3. Radke *et al.* (1978) suggested that elevated ozone concentrations in the plumes from forest fires may be due to the NO_x/NO ratio being higher in the plumes than in the ambient air. However, the ozone concentrations measured in the plumes of the prescribed burns discussed in the present paper were higher than those required to be in equilibrium with the measured ratios of NO_x/NO ; consequently, the observed levels of NO_x and NO are insufficient to account for the elevated ozone concentrations observed in the plumes. The correlations between the high ozone concentrations and the ultraviolet intensities (Fig. 11) indicate that the ozone in the plumes was produced by photochemical reactions. Evidence for the production of ozone by photochemistry in the plumes produced by the burning of forest and grassland slash in Australia has been reported by Evans *et al.* (1977). However, these workers concluded that the photochemical reactions were essentially complete within a transport time of 1 h. Our observations show

Table 1. Measured values of average ozone concentration, ultraviolet (u.v.) values and particle loading in the plumes of the three prescribed burns of forest slash

Date	Local time, h, min	Distance downwind of burn, km	Altitude, m, MSL	Concentration of ozone in the plume in excess of ambient,* ppb	u.v. flux, $W m^{-2}$	Mass concentration of particles in the plume in excess of ambient,† $\mu g m^{-3}$
19 July 1978	1429	3	1676	19.0	28.1	277
19 July 1978	1433	3	1829	16.0	29.2	411
19 July 1978	1438	3	1981	11.0	30.1	282
19 July 1978	1443	3	2134	1.0	24.8	609
19 July 1978	1448	3	2286	10.0	24.8	877
19 July 1978	1454	3	2438	9.0	30.8	293
19 July 1978	1522	10	2682	44.0	23.0	795
19 July 1978	1543	10	2438	29.0	17.2	663
19 July 1978	1539	10	2286	30.0	21.6	663
19 July 1978	1546	10	2134	6.5	17.3	436
19 July 1978	1551	10	1981	43.6	25.6	461
19 July 1978	1557	10	1829	-5.8	14.0	615
19 July 1978	1601	10	1676	-8.1	8.6	461
19 July 1978	1606	10	1524	-2.8	3.1	404
19 July 1978	1609	10	1372	-8.4	1.8	152
19 July 1978	1612	10	1219	-4.0	6.8	401
19 July 1978	1615	10	1067	-11.3	4.3	Not available
19 July 1978	1643	25	2438	40.0	20.9	203
19 July 1978	1648	25	2591	30.0	21.0	67
22 September 1978	1314	6	2438	-18.0	0	Not available
22 September 1978	1318	6	2590	-30.0	1.3	Not available
22 September 1978	1324	6	2438	2.0	27.2	222
22 September 1978	1334	7	2134	-20.4	4.6	Not available
22 September 1978	1405	7	1981	0.5	21.8	725
5 October 1978	1153	1.8	1219	2.9	21.4	190
5 October 1978	1202	1.8	1341	4.8	23.2	246
5 October 1978	1211	1.8	1067	1.0	17.9	285
5 October 1978	1226	9.3	1219	11.9	22.3	12
5 October 1978	1231	9.3	1341	1.4	25.1	38
5 October 1978	1236	9.3	1067	4.6	21.5	65
5 October 1978	1243	9.3	914	7.2	19.0	44
5 October 1978	1254	18.5	1219	15.8	24.5	75
5 October 1978	1300	18.5	1341	7.6	23.9	69
5 October 1978	1309	18.5	914	0.4	22.3	Not available
5 October 1978	1539	1.8	1219	3.9	14.4	210
5 October 1978	1543	1.8	1402	3.1	13.8	693
5 October 1978	1550	1.8	1067	-5.8	8.1	890
5 October 1978	1558	9.3	914	3.4	9.5	50
5 October 1978	1606	9.3	1158	20.9	9.2	490
5 October 1978	1613	9.3	1402	20.0	11.0	142
5 October 1978	1629	1.8	1067	-15.3	2.7	1611
5 October 1978	1647	18.5	1280	14.0	7.1	510
5 October 1978	1652	37.0	1280	16.2	6.7	112
5 October 1978	1700	55.6	1280	26.0	7.5	54
5 October 1978	1709	74.0	1280	2.5	5.8	Not available

* Ambient ozone values ranged from 60–85 ppb on 19 June (generally decreasing with altitude), about 50 ppb on 22 September, and about 60 ppb on 5 October.

† Ambient particle mass loadings were generally $< 10 \mu g m^{-3}$ on 19 July and 22 September and $< 1 \mu g m^{-3}$ on 5 October.

considerable ozone production for travel times between 15 min and 1 h but, as can be seen from Table 1, ozone was still being generated at longer travel times. For example, the measurements on 5 October 1978 at 1647, 1652 and 1700 PDT indicate that the ozone concentration was still increasing at 55.6 km downwind, even though the plume had diluted considerably at this range (as indicated by the reduced particle loadings).

7. SUMMARY OF CONCLUSIONS

Prescribed burns of forest fuels can produce substantial increases in the concentrations of ozone downwind of the burns: ozone concentrations near the tops of the plumes sampled in this study reached values as high as 44 ppb above ambient values. The higher values were generally associated with high u.v. in-

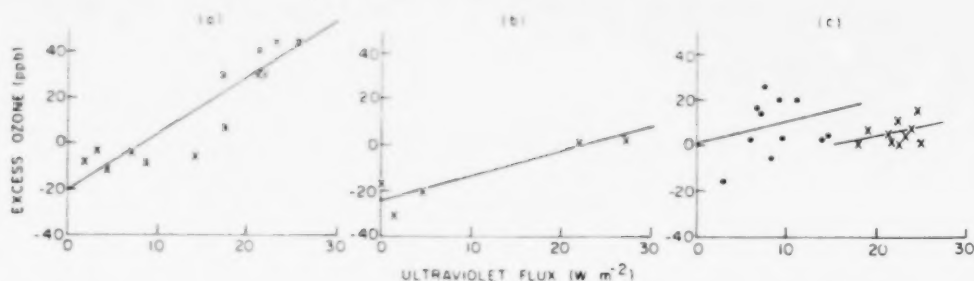


Fig. 11. Excess (above ambient values) of the ozone concentrations in the plumes from the burns against the ultraviolet flux for (a) 19 July 1978 at downwind distances of 10 and 25 km, (b) 22 September at downwind distances of 6 and 7 km, and (c) 5 October prior to 1530 PDT (crosses) and after 1530 PDT (dots).

tensities. Ozone was formed even after plume travel times greater than 1 h.

The prescribed burns also produce a substantial impact on visibility. Neglecting the effects of light absorption, the visibility in the densest regions of one of the plumes was only about 3% of that in the background air.

Particle volume distributions in the plumes from the burns peaked at $\sim 0.3 \mu\text{m}$ where over 80% of the particulate volume resided. A smaller peak at $\sim 0.5 \mu\text{m}$ was also observed. The origin of these peaks is unknown, except that they result from the emission of primary particles by the fires. The burns produced particulate mass at rates of $0.1\text{--}15 \text{ kg s}^{-1}$.

Estimates of particle emission factors from the burns ranged from 0.2 to 2.0%. However, these results are somewhat uncertain, since they assume that the burns were in steady-state during the period of the measurements; also, they are based on fuel consumption rates estimated by foresters rather than being measured precisely. A promising alternative method for determining particle emission factors is through simultaneous airborne measurements of the concentrations of CO_2 and particles in a plume.

The burns that consumed their fuel most rapidly had the highest emission factors, and the fuels with the highest water content produced particles with the highest density. However, since only three burns were investigated, further studies are required to determine if these results are generally true.

The range of emission factors determined in this study are similar to those reported by Butcher and Sorenson (1979) for emissions from wood stoves under various conditions of fire draft. Those values are slightly lower than the value of 3% used by Packham and Vines (1978) for estimating visibility reduction due to prescribed burns.

The burns were substantial sources of cloud condensation nuclei (CCN); $10^{10}\text{--}10^{11}$ CCN were produced

per gram of wood consumed. When these emissions are ingested into a cloud, they produce unusually high concentrations of small ($< 10 \mu\text{m}$ diameter) droplets. This distortion of the cloud droplet size distribution may affect the formation of rain.

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REFERENCES

- Butcher S. S. and Sorenson E. M. (1979) A study of wood stove particulate emissions. *J. Air Pollut. Control Ass.* **29**, 724–728.
- Evans L. F., Weeks I. A., Eccleston A. J. and Packbain D. R. (1977) Photochemical ozone in smoke from prescribed burning of forests. *Envir. Sci. Technol.* **11**, 876–900.
- Forrest J. and Newman L. (1977) Further studies on the oxidation of sulfur dioxide in coal-fired power plant plumes. *Atmospheric Environment* **11**, 465–474.
- Gillani N. V., Husar R. B., Husar J. D., Patterson D. E. and Wilson W. E., Jr. (1978) Project MISTT: kinetics of particulate sulfur formation in a power plant plume out to 300 km. *Atmospheric Environment* **12**, 589–598.
- Hobbs P. V., Hegg D. A., Eltgroth M. W. and Radke L. F. (1979) Evolution of particles in the plumes of coal-fired power plants. I: deductions from field measurements. *Atmospheric Environment* **12**, 935–951.
- Packham D. R. and Vines R. G. (1978) Properties of bushfire smoke: the reduction in visibility resulting from prescribed fire in forests. *J. Air Pollut. Control Ass.* **28**, 790–795.
- Radke L. F. and Hobbs P. V. (1976) Cloud condensation nuclei on the Atlantic seaboard of the United States. *Science, N.Y.* **193**, 999–1002.
- Radke L. F., Stith J. L., Hegg D. A. and Hobbs P. V. (1978) Airborne studies of particles and gases from forest fires. *J. Air Pollut. Control Ass.* **28**, 30–34.
- Veretennikov V. V., Kozluy V. S., Naats I. E. and Ya Fadeev V. (1979) Optical studies of smoke aerosols: an inversion method and its applications. *Optics Lett.* **4**, 411–413.